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RAMAN AND INFRARED SPECTRA OF POLLUCITE *

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ABSTRACT

Raman light scattering, infrared reflection, and infrared absorption spectra are reported for the mineral pollucite and other cesium aluminosilicate formulations of interest to the beneficial use program of Sandia Laboratories. In addition, Raman spectra of crystals of a pure phase pollucite are presented. Assignment of the active normal modes is made; a resonance at 200 cm^{-1} is tentatively assigned to a Cs cage mode; the remaining resonances are assigned to oxygen stretching vibrations of the Al O and Si O bonds, external vibrations of the aluminosilicate framework, and to more complex vibrations involving all of the atoms of the crystal. It is shown that by using a reference crystalline spectrum, other cesium aluminosilicate structures can be identified as polycrystalline in nature with a makeup similar to pollucite. Structural alterations (broken bonds) as well as state changes (crystalline to amorphous) can also be ascertained by Raman spectroscopy. Water is found only in mineral pollucite, where it exists as a monomer, hydrogen bonded to oxygen atoms that form its cage. Satisfactory results were obtained from a range of materials; black to milky to clear in appearance.

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INTRODUCTION

Raman and infrared spectroscopy have been applied to the examination of various cesium aluminosilicates, all similar to the mineral pollucite. The samples examined in this study were furnished by J. K. Johnstone/5846. Sandia Laboratory's interest in pollucite is based on the ability of this material to take up proportionately large quantities of Cs. Sandia is planning to exploit this property and use a pollucite-like material to incorporate (radioactive) ^{137}Cs, a nuclear reactor byproduct. This material can then be used as a gamma-ray source for beneficial use programs.[1]

Our choice of Raman and infrared spectroscopy to examine these samples was motivated by the ability of these techniques to determine atomic structures. These spectroscopies detect vibrational modes that involve the atomic constituents, and these modes in turn can be related to the structure of the material. In addition to the basic structure of the material, atomistic defects and molecular inclusions in the structure can be observed. Examples of these types of species are broken bonds and water. Structural information is useful in at least three ways: (1) structural differences resulting from the method of preparation can be found; (2) structural changes as a function of some interaction, say with gamma radiation, can be assessed; (3) precusors to or the onset of gross mechanical change may be seen. In this last situation, for instance, soft modes that precede the occurrence of solid state phase changes have been observed with Raman spectroscopy.[2] Ultimately, information about atomic structure can be related to the mechanical behavior or integrity of the material.

With these goals in mind we have developed a three point program to study pollucites: (1) to obtain the "reference" Raman and infrared spectra and to correlate them with the structure of pollucite; (2) to simulate the gamma-ray dose to the material and/or the effects of the transmutation of ^{137}Cs;[3] (3) ultimately, to work directly with the radioactive material.

At this point in the study, most of the effort to complete objective (1) has been done. This memo will be devoted principally to the discussion of these results. Some experiments with radiation simulation will be mentioned briefly. The objective of working with hot material will be discussed in the proposed program portion of this paper.

We have included discussions of sample preparation, pollucite crystal symmetry, Raman spectra of crystalline, mineral, and ceramic forms, a crystal-to-glass comparison, IR spectra, water in pollucite, radiation simulation experiments, conclusions, and a proposed program.

POLLUCITE SAMPLES

Three candidate samples of cesium aluminosilicates for ^{137}Cs incorporation, all similar to pollucite, were examined (See Table I). These were prepared by J. K. Johnstone, 5846. Each of these three had a slightly different composition. Samples H-260 and H-275 were prepared from reagent-grade chemicals, while H-273 was manufactured from CsOH Montmorillonite clay. All were prepared in the same manner: hot pressing a dehydrated powder near 1000°C. These samples are shown in Figure 1, where H-275 is milky colored, H-260 is partially translucent, and H-273 is black. Raman and infrared reflection spectra were successfully taken from all these materials. Raman light scattering from H-273 and H-275 was limited to the near-surface region.

Two other sample types known to have pollucite formulations were also examined. One was a mineral (See Figure 1 and Table I for the chemical formulation) and as such was polycrystalline. The others were laboratory-grown, hydrothermal crystals obtained from S. A. Gallagher and G. McCarthy of Pennsylvania State University through J. K. Johnstone. Some of the small crystalline samples on a fixture used for light scattering are shown in Figure 2.

In this work we correlate Raman scattering and infrared spectra with the known crystalline structure of pollucite a. . look for variations in other cesium aluminosilicate structures based on change in these reference spectra. Pollucite has cubic symmetry - O_h^{10} (Ia3d).[4] Table I lists the stoichiometry of the mineral pollucite and of the pure-phase crystals (the hydro-thermal crystals). Other alkali oxides, principally Na_2O, are present in the pollucite mineral. Also, mineral pollucite is known to be a zeolite with four

water molecules per unit cell. In the case of the pure-phase material, no other alkali oxides are present, and there is no water. In both cases, however, the space group is O_h^{10} (Ia3d).[4,5]

Two dimensional projections of four unit cells of pollucite are shown in Figure 3.[4] An aluminosilicate framework is the basis of the pollucite structure. The silicon and aluminum are fourfold coordinated with the oxygen atoms, forming tetrahedra that are the basic building blocks of the framework. The structure is composed of 4, 6, and 8 member rings of tetrahedra. The Si and Al are randomly placed at the tetrahedral sites in the crystal. To establish the fourfold valency of aluminum, an electron is garnered from an alkali atom. Consequently, in all the aluminosilicate structures with which we will be dealing, the Cs_2O to Al_2O_3 ratio is 1:1. In the aluminosilicate framework, sixteen large cages are formed in each unit cell. These have diameters on the order of 6.8 Å and contain Cs or H_2O molecules. In what follows, we shall be interested in finding the modes that can be characterized with the movements of the bridging oxygen atoms between tetrahedra, modes of the tetrahedral framework, and modes that predominantly involve Cs.

It should be noted that two other crystalline formulations of cesium aluminosilicate have been found. These are $Cs_2O \cdot H_2O \cdot 2SiO_2$[6] and $Cs_2O \cdot H_2O \cdot 10SiO_2$,[7] and neither is cubic. In all of our spectral results to date we have seen no evidence of a material that has other than cubic symmetry.

RAMAN SPECTRA OF CRYSTALLINE POLLUCITE

The first step in our program has been to obtain and understand the spectra of the pure-phase pollucite crystals as a stepping-stone for interpreting

structural alterations that might occur in other cesium aluminosilicates. We are most fortunate to have crystals of the pure-phase material and a sample of the mineral to examine. As mentioned, both materials have a space group symmetry of O_h^{10}. The crystals are very complex and have a large number of atoms in the primitive cell, resulting in some 250 normal modes for the pure-phase crystal. Not all, however, are either Raman or infrared active, as shown in Table II. Also shown in Table II are the polarizability tensors and infrared activities of appropriate modes. Because the irreducible symmetry inversion operator is one of the space group operations of O_h^{10}, the active modes are either Raman active or infrared, but not both.

Theoretically, the A, E, and F modes (the singly, doubly and triply degenerate modes) that are Raman active can be distinguished. However, this resolution necessitates alignment of the crystallographic axes with the scattering axes of the equipment. Clearly, this procedure cannot be done in the case of the polycrystalline samples. Unfortunately, the pure-phase crystals were very small polyhedra clumped together with dimensions $\sim 150\mu m$; consequently we have not yet been able to align the crystallographic axes with the scattering axes of the crystals. Nevertheless, the A modes can be separated from the E and F modes; the A modes have a diagonal polarizability tensor, and therefore, under any coordinate axis rotation, the scattering from them will remain polarized. This property is not the case for the E and the F modes; they will present both polarized and depolarized spectra.

Table II also lists the atoms that participate in the various symmetry modes. The normal modes are built up from the symmetry modes of the same irreducible character. The interesting point is that for the Raman-active modes, only

those modes of E or F character will contain a mode that involves the motion of Cs. Further, the number of these is limited to 2.

A comparison of the polarized and depolarized spectra from pure-phase pollucite is shown in Figure 4. A list of the identifiable resonances seen in pure-phase pollucite is in Table III, along with the assignment of the appropriate irreducible symmetries. Due to the crystal complexity no attempt has been made to determine completely the atomic contribution to all of the normal modes of pollucite. A qualitative statement about the atomic character of these modes can be made, however. The band at 1100 cm^{-1} is the oxygen stretch of the SiO bond. This band has been identified with the oxygen stretch of the Al-O bond.[9]

Both of these bands are composites of several normal resonances involving oxygen movement. A series of sharp and symmetrical resonances dominate the spectrum at lower frequencies from about 498 to 70 cm^{-1}. All of these except one, we believe, are vibrations of the aluminosilicate tetrahedra in the crystalline framework. The one exception is the band at 200 cm^{-1}. We will reserve discussion of this mode to the comparison of the mineral with the pure-phase material. Finally, a series of resonances has been detected between 500 and 1000 cm^{-1}, and these are complex vibrations involving all of the atoms of the unit cell.

A comparison of the Raman spectrum from the mineral and that from the pure-phase material is shown in Figure 5. Resonances seen in the mineral are listed in Table III. The spectra from the two materials are remarkably similar. The oxygen stretching bands are in agreement, as are modes associated with the external vibration of the framework. Furthermore, when the weak bands in the

material could be detected they too agreed. The mode at 200 cm^{-1}, however is very weak in the mineral in comparison to the pure-phase material. In the crystal spectrum this mode was assigned to the irreducible symmetry of E or F. Now in the mineral there are 12 atoms of Cs per unit cell instead of the 16 that exist in the pure-phase crystal. In the positions of the missing Cs atom are water molecules. The weakening of the mode is probably due to the loss of translational regularity of the Cs atoms, implying that this mode involves the participation of Cs. Supportive of this assignment is the observation that in the Argon matrix formation of CsO_4, low frequency modes near 200 cm^{-1} have also been observed. Thus, 200 cm^{-1} resonance most likely involves Cs atom participation.

RAMAN SPECTRA OF CERAMIC CESIUM ALUMINOSILICATES

Two of the SLA-prepared materials appeared to be ceramic or polycrystalline materials. This conclusion is rather straightforward when the Raman spectra of the ceramics are compared with that from the crystals (See Figure 6), and also when the resonant frequencies are compared (See Table III). In Figure 6, spectra from the crystal pollucite, the mineral pollucite, and the pollucite samples H-275 and H-273 are displayed. There is good agreement among most of the resonances in frequency position, relative intensities, and line shapes. From this general agreement it can be inferred that their basic structure is very similar. In addition, from the fact that most of the lines are symmetric and sharp, it can be argued that the scattering is occurring from a system of highly regular order, i.e., crystals. Finally, the widths of these symmetric lines indicates that the crystalline sizes are at least on the order of the wavelength of light, 5000Å.

There are some differences amongst these spectra, as can be seen in the frequency regime near 1000 cm^{-1}. Here one material (H-273) exhibits a different behavior: an extra band appears at 973 cm^{-1}, and there is weakening of the bands at 1000 and 1100 cm^{-1}. A possible explanation for this behavior is the disruption of the Si-O-Si or Al-O-Si bonds joining tetrahedra. The bands at 1050 and 1100 cm^{-1} previously were associated with this bridging. Other studies have identified a band near 973 cm^{-1} in silicate materials as being due to the oxygen stretch of a dangling or non-bridging Si-O bond.[10, 11] Lattice disruption - resulting in dangling oxygen bonds - might be expected at that sample, because it was made from Montmorillionite clay and CsOH. The clay contained a large percentage of impurities which, when incorporated into the material, could break oxygen bonds connecting tetrahedra. (See Table II.) Another interesting observation, based on the comparison of Raman spectra, is that in going to the silica-rich formulation of cesium aluminosilicate (H-275) the basic structure does not seem to have been altered much from that of pollucite. Apparently the basic aluminosilica tetrahedral framework is retained, with proportionally more silicon atoms than aluminum atoms in the H-275 formulation. This premise is also in agreement with the observation that the band at 1000 cm^{-1} (the Al-O stretch) in comparison to the band at 1100 cm^{-1} (the Si-O stretch) is weakened in the case of H-275 as compared to the pure-phase pollucite. Note also that the band at 390 cm^{-1} is strengthened relative to the band at 300 cm^{-1} in increasing the Si content in H-275 from the crystalline material. This result suggests that the mode at 390 cm^{-1} involves Si, while that at 300 cm^{-1} involves Al.

In Figure 7, a comparison of the Raman spectra from sample H-260 and crystalline pollucite is shown. In contrast to the similarities discussed before, now significant differences between the spectra can be noted. New Raman-active modes have appeared, bands have broadened, and the symmetrical shape of the bands has been lost. All these features highlight the conclusion that there is a difference in the state of the material, and it can be shown that this difference arises because the H-260 sample is amorphous.

Included in Figure 7 is a Raman spectrum of a vitreous silica, Suprasil 2. There are a number of notable similarities between it and that of H-260; for instance, the broad structureless scattering below 400 cm^{-1}, and the alignment of a number of peaks. This comparison underscores our conclusion that H-260 is amorphous. Additionally, there have been reports of Raman spectra of other ternary systems, similar to the one that we are dealing with, which were known to be amorphous. Figure 8 is a reproduction of spectra obtained from sodium borosilicate glasses, about which the author concluded that he was dealing with a glassy system, predominately SiO_2 in nature.[12] There is a remarkable similarity between the spectrum labelled 3 and that obtained for H-260, which further strengthens our conclusion that H-260 is a glassy material.

Some additional conclusions about the character of various resonances can be made in intercomparing the three spectra in Figure 7. Vitreous silica, in one important feature, is very much like the aluminosilicate materials: it i made up of interconnected tetrahedra. In the case of silica they are all S tetrahedra, and - in the vitreous state - randomly connected. Now note that

in both cesium aluminosilicates there is a band near 300 cm^{-1}, which does not occur in vitreous silica indicating that this mode involves Al. A band near 400 cm^{-1} exists in all these spectra. This band previously has been identified as involving Si[13]; it is appropriate in the case of pollucite to do so also. Furthermore, in silica the band at 800 cm^{-1} is known to be due to oxygen in plane-stretch of the Si-O-Si bond.[13] Again, we will make this assignment in the cesium aluminosilicates. We are left with two bands that are coincident in these materials that have not been satisfactorily identified in vitreous silica: those at 480 and 600 cm^{-1}. We tentatively suggest that the mode at 400 cm^{-1} is an external vibration of the framework with the tetrahedra moving as units, and the 600 cm^{-1} mode quite possibly depends upon some local coordination of atoms in neighboring tetrahedra.

IR SPECTRA OF POLLUCITE AND CERAMIC CESIUM ALUMINOSILICATES

By far the simplest method of obtaining IR spectral data for thick, insulating materials (such as the present pollucite samples) is to examine the material by specular reflection of a near-normal indicent beam. Unfortunately, large changes in the refractive index across vibrational bands distort the band shapes and intensities relative to transmission IR measurements and Raman spectra. To avoid this difficulty we proceeded by the well-known technique of suspending a few milligrams of material in an IR-transparent KBr pressed disc.

These absorption spectra from the region 400 to 1500 cm^{-1} are presented in Figure 9. Reflection spectra from 100 to 1500 cm^{-1} are shown in Figure 10. From a comparison of observed Raman and IR frequencies in Table III, the non-correlation of modes for the two techniques, as predicted by the selection

rules for the crystalline materials, is confirmed. In agreement with the Raman spectra, the IR bands for the pure-phase material appear slightly broader than for the crystalline materials, consistent with the presence of an amorphous phase. This broadening is most noticeable in the bands between 700 and 800 cm^{-1}; however, the effect is not nearly as apparent as in the Raman spectra.

The intense band around 1030 to 1050 cm^{-1} for all samples is composed of a number of modes; its center is shifted down considerably from the analogous peak of 1090 cm^{-1} observed in vitreous silica, but correlates well with IR spectra for other aluminosilicates. (Kaolin clay, for example, has a broad, multistructured band in this region with a maximum intensity at 1035 cm^{-1}.[14]) The peaks near 760 to 780 cm^{-1} correspond to a peak in vitreous silica at 790 cm^{-1}, while peaks from 750 to 625 cm^{-1} correspond to observed bands in alumina (Al_2O_3).

The very intense band observed in all the reflection spectra from 445 to 455 cm^{-1} is observed in both Al_2O_3 and SiO_2. The remaining observed IR band is a sharp peak seen in the reflection spectra at 156 cm^{-1} in all cases. While there are bands observed at 150 and 170 cm^{-1} for SiO_2 and Al_2O_3, respectively, these are very weak features compared to the present data. Although it is conceivable that the feature at 156 cm^{-1} could be related to Cs atom motion, a more positive assignment must await definitive experiments with similar aluminosilicate systems not containing Cs.

WATER IN POLLUCITE

A search was conducted for water in all of the pollucite materials. In the glass, ceramic, and pure-phase crystalline pollucite materials, no detectable quantity of OH was found in either Raman or IR spectra.

However, water is known to be abundant in the mineral pollucite (See Table I). Indeed, we found vibrations that can be identified with the water molecule both in the infrared and Raman spectrum of the mineral. (See Figures II & I.). The frequency peaks of the water resonances are in Table III, where they are compared with those of an isolated water molecule.[15] In agreement with the isolated water molecule, the three peaks seen in pollucite have the correct Raman and infrared activities and have similar vibrational frequencies. From this identification we conclude that water in pollucite is a monomer. It is known that water in a liquid polymerizes, and that this polymerization dra stically alters the Raman spectra from that of the isolated molecule.[6] Such alterations have not been seen here, supporting our conclusion that water is in pollucite as individual molecules, monomers. Finally, it is most probable that the monomeric water molecule resides in the large oxygen cages in the aluminosilicate framework referred to earlier. However, for water in pollucite - in comparison to isolated water vapor - there has been a down shift in frequency, line broadening, and the formation of low frequency tails in the symmetric and asymmetric stretching bands. We believe that these effects are caused by hydrogen bonding of the water molecule to the oxygen atoms that form its cage.

RADIATION SIMULATION STUDIES

Two preliminary tests have been performed to simulate radiation effects in pollucite. In the first, the three SLA samples of pollucite were irradiated with ^{60}Co gamma-rays at the LLL radiation facility to a dose of 1×10^{9} rads. In the post-test examination no alteration of the Raman spectra was observed. Another radiation test was tried with inconclusive results. Samples H-273 and H-275 were irradiated with a 50 keV electron beam, to a total dose of

$\approx 10^{12}$ rads. Post test examination again revealed no spectral alteration, although this conclusion could be misleading for the following reasons. The e beam radia a was believed to be deposited to a depth of about 10 μm. Due to absorption of light in the H 275 sample and light scattering in the H 27% sample, the portion of the material sampled by Raman scattering was also confined to a region near the surface. Unfortunately, the exact depth of penetration of the light beam is not well known in either case. In the irradiated samples, Raman scattering could have been probing to a depth significantly greater than the e beam penetration. In this case the signal from the unirradiated volume of material could then obscure the signal from the (relatively small) irradiated portion. On the basis of this single experiment it cannot be safely concluded that the pollucite structure was unaffected by the high level of radiation.

CONCLUSIONS ABOUT THE USE OF RAMAN AND INFRARED SPECTROSCOPY

The Raman spectra of crystalline pollucite are now known. These will serve as the base (fingerprint) spectra, which can be used to identify crystalline pollucite and to assess structural alterations.

Crystalline forms can be distinguished from amorphous forms of pollucite. Thus, changes of state in a material that might affect mechanical integrity can be observed, e.g., radiation-induced devitrification.

Successful examination of materials from clear to black and from the size of micrometers to centimeters has been accomplished.

SUGGESTED PROGRAM

A program is proposed to accomplish the three objectives outlined above: the characterization of pollucite, radiation simulation, and working with radioactive material.

A. Characterization of Pollucite

Definite progress has been made in identifying and unravelling the Raman and IR spectra of crystalline pollucite. Additional effort is needed, however, to verify the modes associated with Cs. Two approaches will be taken. The first will involve aligning the crystalline pollucite with the optical axis of the scattering experiment allowing the separation of the E_{1g} and E_{2g} modes. The second is to compare the Raman spectra of pollucite to analcime, which has the identical crystal structure to pollucite, but does not contain Cs.[4] The appropriate Raman spectra must be obtained in the laboratory.

Further characterization will be necessary of the form of pollucite selected to be used in the beneficial-use program. Raman and infrared spectroscopies, as demonstrated, could be used to obtain important structural information about these materials. Thus, Raman and IR measurements should be included in the studies to develop pollucite-like materials for the beneficial-use program.

B. Radiation Simulation

Pollucite could receive doses approaching 10^{13} rads during the proposed 90 year source lifetime. Possible ways of simulating this dose include ^{60}Co and e-beam irradiation. Presently very hot ^{60}Co sources

are available at the Savannah River Plant, Aiken, South Carolina. Use might be made of this facility. Furthermore, (as discussed above), initial attempts with e-beam radiation have already been tried. It is proposed that these experiments be expanded, in conjunction with determinations of the optical skin depth of the appropriate materials.

Another method to achieve radiation deposition alone (without transmutation) would be to make use of the Cs-pollucite sources planned for fabrication at Oak Ridge. Here the base material, not incorporating the radioactive Cs, could be placed around the hot sample.[18] These samples could be periodically removed and examined in the laboratory.

Finally, it is known that a major effect on the crystal structure could come about through the transmutation of Cs^{+1} to Ba^{+2}. This valence change could disrupt the crystal structure. Neutron irradiation experiments have been used before to simulate transmutation, for example, the conversion of boron to helium in borosilicate glasses.[19] Such neutron-generated transmutation effects might be used to simulate valency changes in pollucite.[20]

C. Examination of Radioactive Material

Remote Raman spectroscopy could be used to examine hot materials. This procedure could be accomplished fairly easily, because only three types of apparatus need be placed in the radiation environment. These would include two lenses - one for focusing the laser into the sample and the other for collecting the scattered light - mirrors to direct the light

beams, and positioning stages. All of the sophisticated equipment, including the laser, spectrometers, and electronics, would be outside the radiation environment. These sampling techniques are already being developed for use with tritium in the Tritium Research Laboratory at SLL. Such a system might be designed for hot cell use at SLA.

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Table 1

Cesium Alumina-Silicate Samples

A. Hot Pressed Powders

$Cs_2O \cdot Al_2O_3 \cdot nSiO_2$

Designation	Physical Appearance	n	Impurities (W/%)
H-260	Transulent	7.5	-
H-273	Black	4.0	6.5
H-275	Milky	6.6	-

B. Crystals

Mineral

$6\ Cs_2O \cdot 2Na_2O \cdot 8\ Al_2O_3 \cdot 32\ SiO_2 \cdot 4\ H_2O$

Pure-Phase

$Cs_2O \cdot Al_2O_3 \cdot 4\ SiO_2$

Table III

Mode Frequencies of Pollucite

Raman

Crystal	A		E+F	E+F	?	A	E+F	A	←——— E+F ———→									
Polarized	69		140	200		297	393	477									1044	1114
Depolarized	69		141	201		297	392	477	559	610	652	711		807	861	920	1016	1108
Mineral	71	115		197	268	297	392	478	520		667						1043	1120
H-273	72		129		277	296	387	472			670					973	1076	
H-275	82	106	133	205	271	300	388	476		597	667	709	771	809	851		1044	1114
H-260		107					383	485						788			1002	1122

Infrared

	F				F	F		F	F	F	F			F	F	F
H-260	155*				384*	455*		595		720	775			1000^S	1050	1180^S
H-273	155*				384*	442*		622	690^S	728	758			995	1040	1190^S
H-275	155*				384*	446*		626	690^S	727	761			980^S	1030	1125^S
Mineral						441		625	695^S	732	763				1032	1120^S

S-Shoulder

* - Reflection Spectra

Water in Mineral Pollucite

Raman 3602

IR 3650, 3545^S, 1627

Table II

Normal Modes for Srystalline Pollucite O_h^{10} (Ia3d)

Character	Raman Polarizability	IR Active	No. of Modes	Symmetric Mode Composition O	Cs	Al & Si
A_{1g}	XX + YY + ZZ		4	✓		✓
E_g	XX + YY + ZZ, 3(XX - YY)		10	✓	✓	✓
F_{2g}	XY, YZ, ZX		14	✓	✓	✓
			28			
F_1		X,Y,Z	16	✓	✓	✓

Total number of normal modes = 250

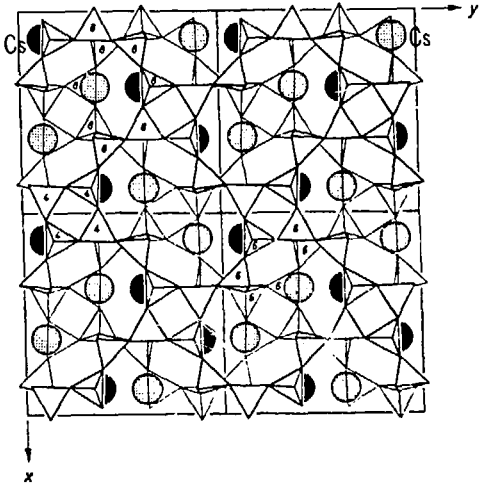

Figure 3. Two dimensional Projection of Four Unit Cells of Pollucite.[4] The solid circles are Cs atoms 1/8 of the side of a unit cell above the bottom face and the shaded circles are 3/8.

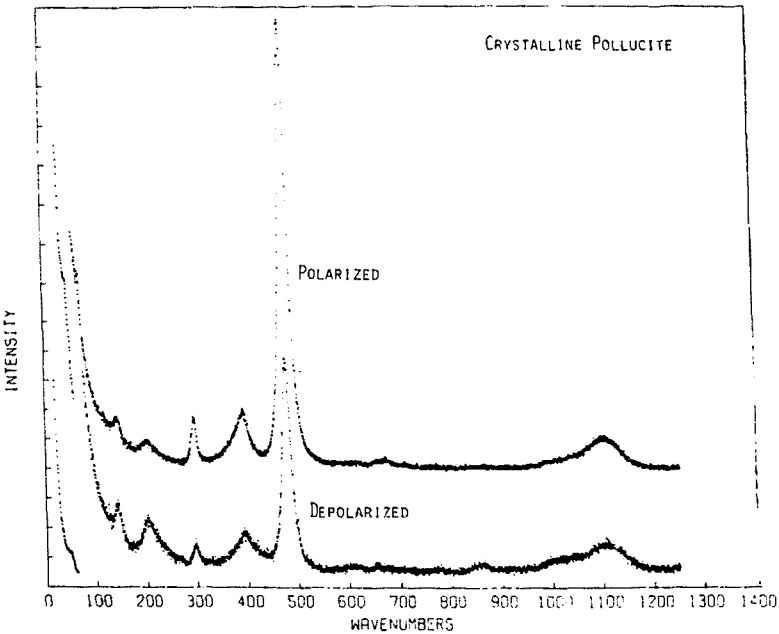

Figure 4. Polarized and Depolarized Raman Spectra of Pure-phase, Crystalline Pollucite.

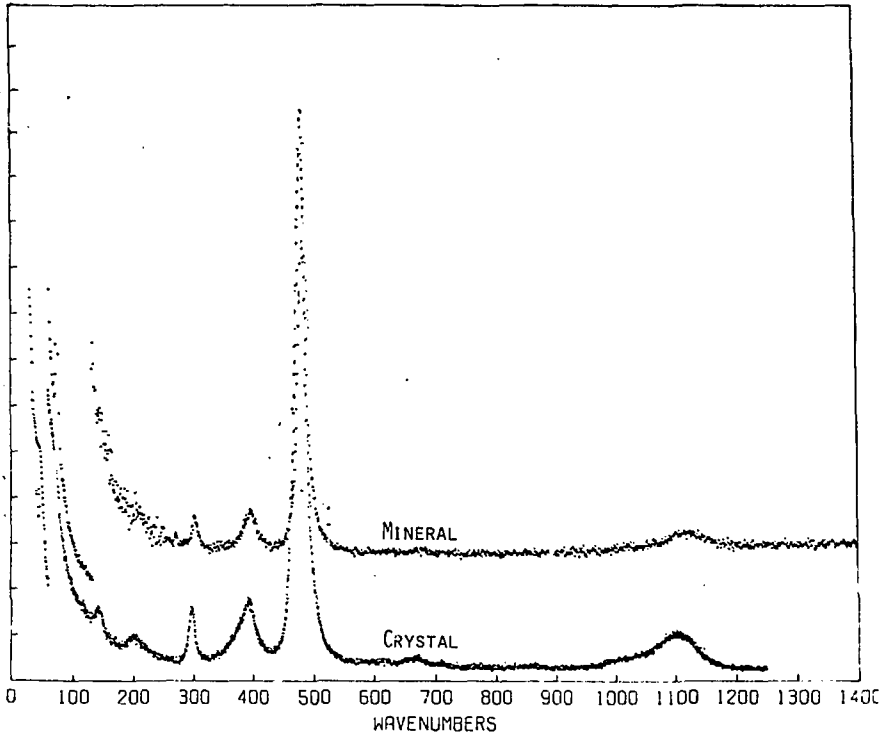

Figure 5. Raman Spectra of Pure-Phase, Crystalline Pollucite and Mineral Pollucite.

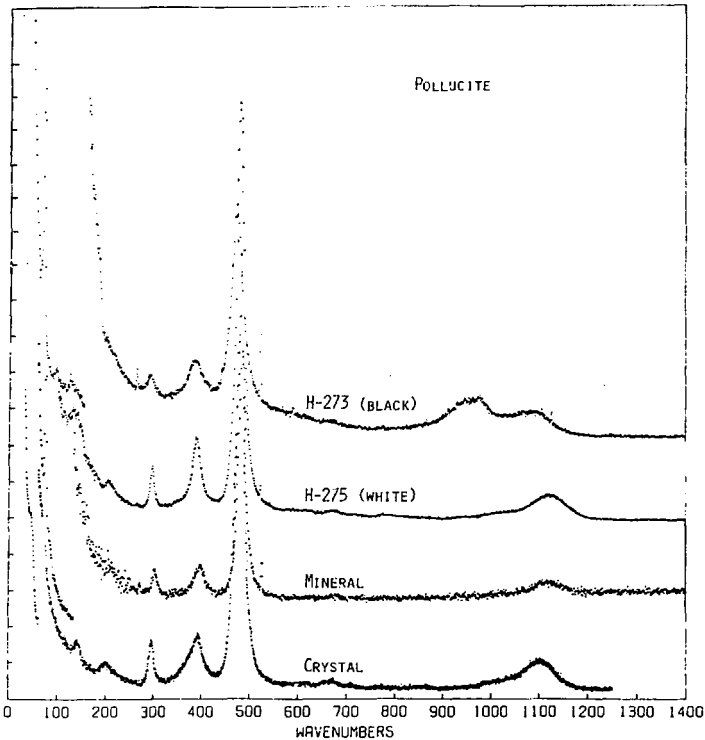

Figure 6. Raman Spectra of Crystalline Forms of Cesium Aluminosilicate Materials. All of the samples examined were crystalline on polycrystalline materials.

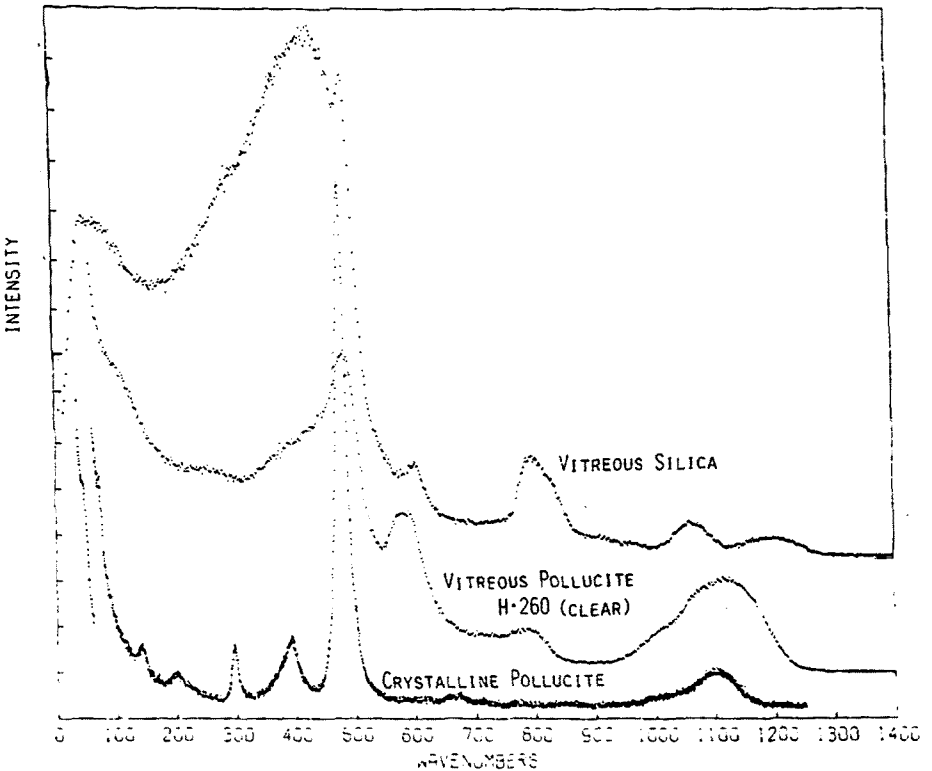

Figure 7. A Comparison of Raman Spectra of Pure-Phase Crystalline Pollucite, Vitreous Pollucite and Vitreous Silica.

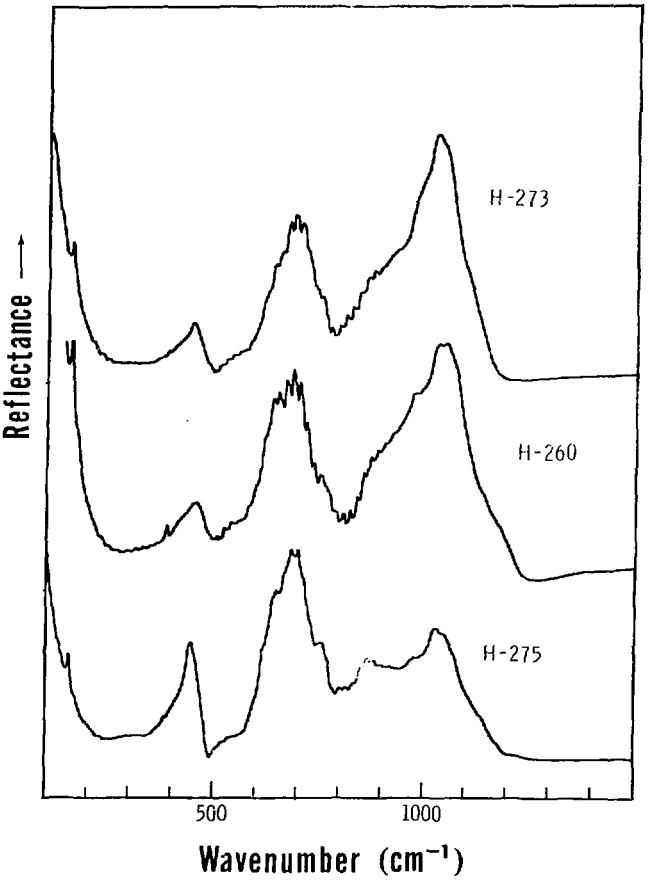

Figure 10. Infrared Reflection Spectra of Cesium Aluminosilicate Materials.

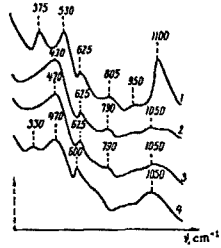

Figure 8. Raman Spectra of Sodium Borosilicate Glasses.[12]

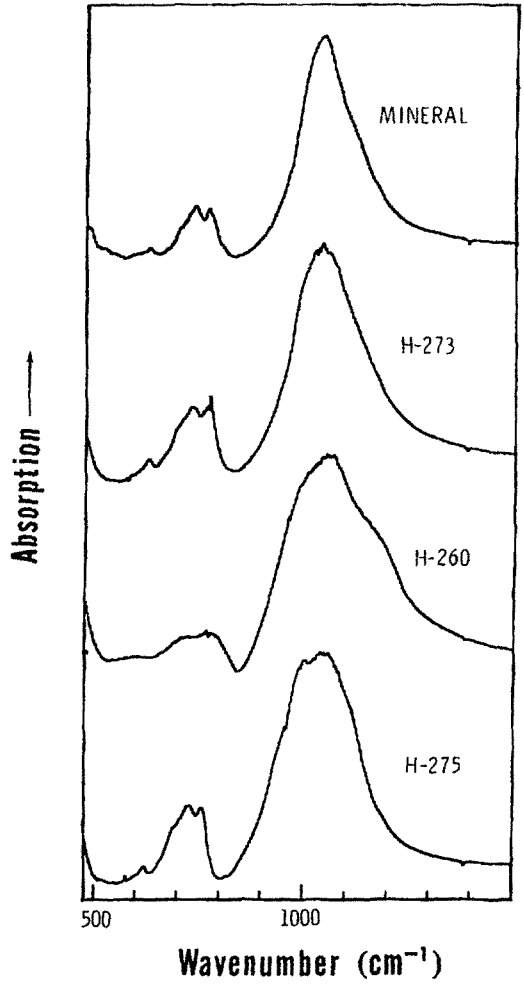

Figure 9. Infrared Absorption Spectra of Cesium Aluminosilicate Materials.

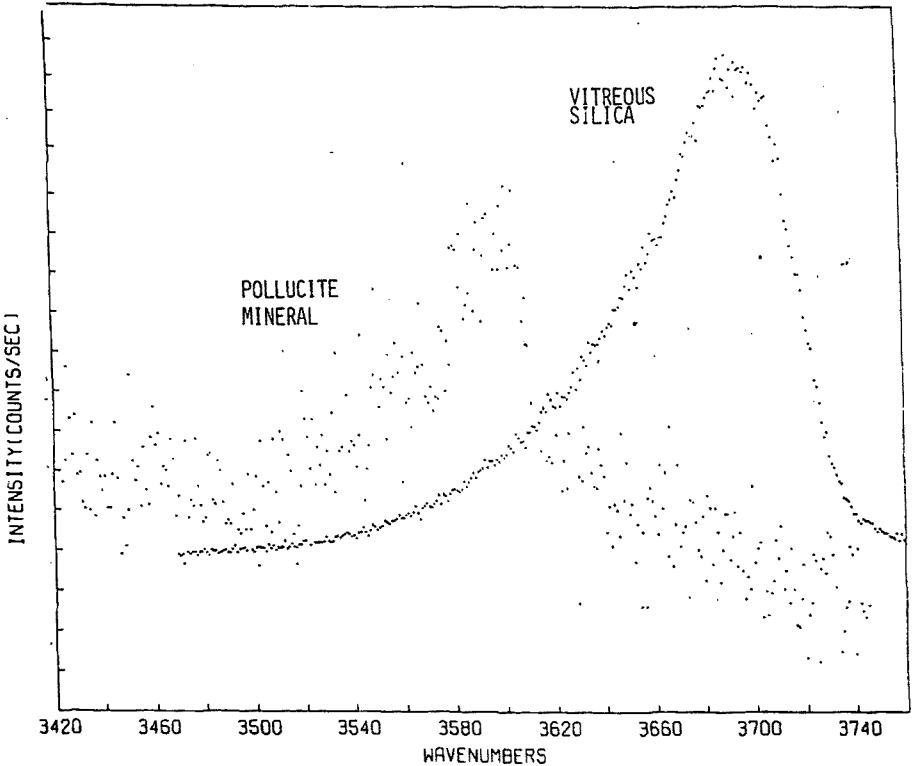

Figure 11. Raman Spectrum of Water in the Mineral Pollucite. The Raman spectrum of hydroxyl in vitreous silica is included for reference. The band observed is due to the symmetric stretch of the triatomic molecule.

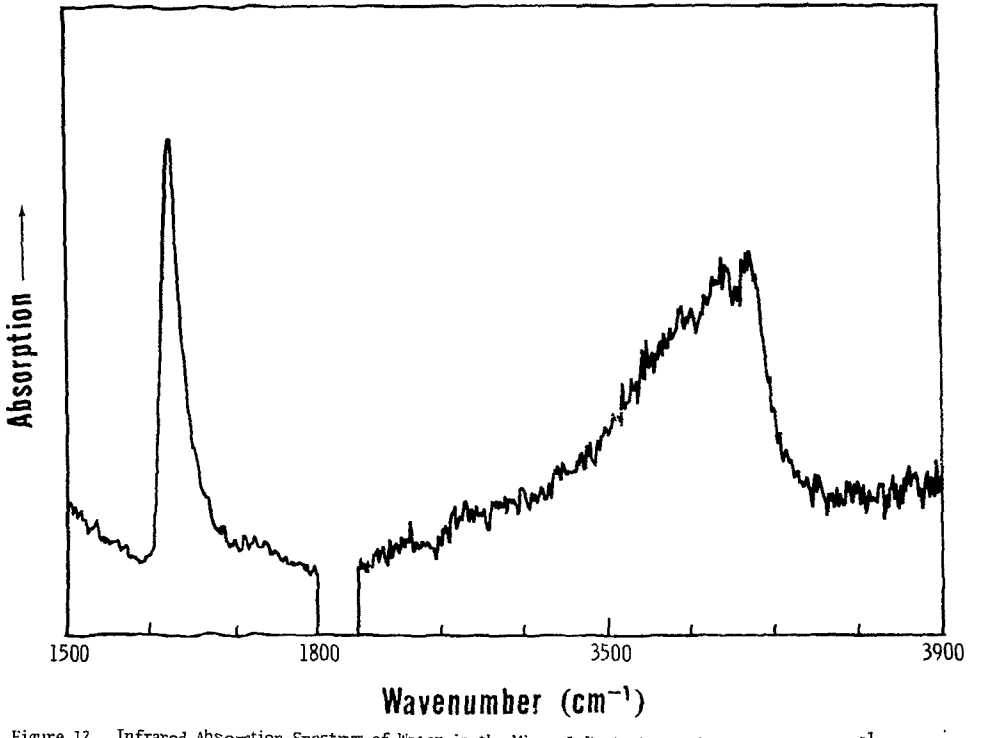

Figure 12. Infrared Absorption Spectrum of Water in the Mineral Pollucite. The band at 3650 cm^{-1} is due to the asymmetric stretch and the band at 1627 cm^{-1} to the bending frequency of the triatomic molecule.